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Fabrication process of YBCO thin film starting from amorphous film for microstrip line device

J. Muyari, N. Kobayashi, S. Takahashi, K. Hayashi, A. Saito*, S. Ohshima

*Graduate School of Science and Engineering, Yamagata University, 4-3-16 Jonan, Yonezawa, Yamagata 992-8510, Japan***Abstract**

We investigated the process for fabricating $\text{YBa}_2\text{Cu}_3\text{O}_y$ (YBCO) thin film microstrip lines from amorphous thin films. First, we made amorphous YBCO thin films and then post-annealing was carried out. The amorphous 300-nm-thick YBCO thin films were deposited on a CeO_2 buffered $\text{r-Al}_2\text{O}_3$ substrate by pulsed laser deposition. The 50-nm-thick CeO_2 buffer layer was made by a facing target sputtering system. We examined the optimal conditions of the post-annealing process by using a two-step crystallization process. The heating rate of the first step was set to 360 K/min until 1023 K, and then the heating rate of the second step was set to 20 K/min until 1073 K. The oxygen pressure was kept to 3000 Pa at 1073 K for 210 min. From the XRD measurement, we observed that the YBCO thin films grew c-axis orientation on the $\text{CeO}_2/\text{r-Al}_2\text{O}_3$ substrate. Both YBCO and CeO_2 ϕ -scan peaks showed four-fold symmetry and rotated 45 degrees between YBCO (102) grain axis and CeO_2 (220) grain axis. The surface roughness, $\Delta\omega$, and $\Delta\phi$ of YBCO thin films were 35.12 nm, 0.89 degree, and 1.51 degree, respectively. We found these to be the optimal post-annealing conditions in which to make a single phase of YBCO films with T_c of 87 K from amorphous YBCO thin films. Therefore, our process is useful to make high-quality YBCO microstrip lines.

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Open access under [CC BY-NC-ND license](https://creativecommons.org/licenses/by-nc-nd/4.0/).*Keywords: amorphous; YBCO thin film; two-step crystallization process; High-temperature superconductor***1. Introduction**

Devices using high- T_c superconducting (HTS) thin films have been increasingly miniaturized because the fabrication technique for HTS thin films has been greatly improved [1]. There are dry-etching and wet-etching as a method for processing HTS thin films [2, 3]. For trying to fabricate fine patterns of HTS microstrip lines, the electron cyclotron resonance (ECR) plasma etching is a typical etching method [4, 5]. Using the ECR plasma etchings, however, the YBCO fine microstrip lines lose superconductivity due to the damage of beam plasma energy. Ion bombardment produces oxygen defects in the edge of YBCO microstrip lines [6]. In fact, etching damages have been frequently observed to degrade superconductivity in YBCO thin films. We developed a process for fabricating YBCO microstrip lines from amorphous thin films. There are some advantages and possibility in this method as described below; the time for depositing an amorphous thin film can be shortened, because of with a high-deposition rate and a room temperature deposition; the etching time for patterning can also be shorter than that in the conventional process; high-quality HTS devices may be realized using this method, since the oxygen deficit of the edge of the HTS patterns by the dry-etching process may be able to decrease. In connection with it, it is effective also in formation of miniaturized HTS devices. We do not yet know the optimal conditions for changing amorphous YBCO thin films into superconducting YBCO thin films with a good crystallinity and high- T_c . Sato et al. have reported producing superconducting YBCO thin film with T_c of 75 K by annealing of amorphous YBCO thin films [7]. In this work, we examined the optimal conditions for the fabrication of the superconducting YBCO thin films starting from amorphous YBCO thin films and characterized the crystallinity and electrical properties of the superconducting YBCO thin films.

* Corresponding author. Tel.:0238-26-3289; fax:0238-26-3289.
E-mail address: atsu@yz.yamagata-u.ac.jp

2. Experimental

We have considered fabricating a microstrip line structure using amorphous YBCO thin films that were patterned by using standard photolithography and dry-etching techniques. We investigated the optimal post-annealing conditions to make a single phase of YBCO thin films from amorphous YBCO thin films, before fabricating microstrip lines. The r-Al₂O₃ single crystal plate was used as a substrate because of low loss tangent, high chemical stability, and mechanical strength. However, we must overcome the problems; for example, the mutual diffusion of Al atoms into the YBCO phase and lattice mismatch between r-Al₂O₃ and YBCO. To solve such problems, many researchers have examined buffer layers [8-10]. We used a 50nm-thick thin film CeO₂ buffer layer on r-Al₂O₃ substrates made by facing target sputtering. For the deposition of CeO₂, we used substrate temperature of 1028 K and a sputtering time of 1 hour with a total gas pressure of 10 Pa and an Ar/O₂ ratio of 32:11. After the sputtering deposition, the CeO₂ was annealed at 773 K for 1 hour in an oxygen atmosphere in the same chamber. The CeO₂ was also post-annealed in the heating furnace for 6 hours at 1323 K in air in order to improve the crystallinity. The surface roughness, $\Delta\omega$, and $\Delta\phi$ of CeO₂ thin films were 1.5 nm, 0.37 degree, and 1.11 degree, respectively. These were sufficient values in order to obtain an epitaxial growth film on the CeO₂ thin films. The amorphous YBCO thin films were deposited on CeO₂/r-Al₂O₃ substrates at room temperature by the pulsed laser deposition (PLD) method. The amorphous YBCO thin films were 300 nm thick. The laser power was fixed at 200 mJ/pulse. The pulse frequency of laser was fixed at 5 Hz. The four annealing conditions in the sintering process were changed with oxygen pressure from 1000 to 10000 Pa, heating rate of the first step from 120 to 600 K/min, heating time from 150 to 210 min and maximum temperature of heating from 1013 to 1073 K, respectively. The oxygen pressure was the most important condition in the four conditions. The crystallinities and T_c values of these YBCO thin films were hardly dependent on the maximum temperature, heating rate, and heating time. The optimal condition was obtained as a result of investigation of the four annealing conditions in the sintering process. Fig. 1 shows a diagram of the optimal post-annealing conditions. We used the two-step crystallization process and oxygen annealing process in the same chamber [11]. For the two-step crystallization process, the heating rate of the first step was set to 360 K/min until 1023 K in order to quickly reach the growing region of the c-axis, and then the heating rate of the second step was set to 20 K/min until 1073 K to reduce an overshoot of the maximum temperature. The oxygen pressure was kept to 3000 Pa at the temperature of 1073 K for 210 min. After the two-step crystallization process, an oxygen annealing process was carried out. The sample was cooled rapidly to 723 K. Then the oxygen pressure was increased to 1 atm and kept there for 120 min.

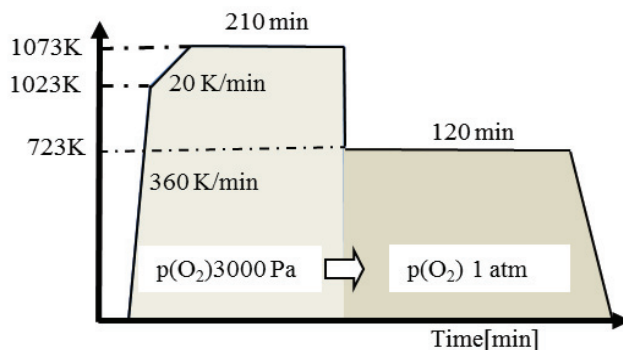


Fig. 1 Two-step annealing program for amorphous YBCO thin films.

3. Results and discussion

3.1. Dependence of the oxygen pressure

The heating rate of the first step was set to 120 K/min until 1003 K, and then the heating rate of the second step was set to 20 K/min until 1053 K. The oxygen pressure was kept to 1053 K for 150 min. The oxygen pressure was changed with 1000 to 10000 Pa. Fig. 2 (a) plots X-ray diffraction (XRD) patterns varying the oxygen pressure. YBCO/CeO₂/r-Al₂O₃ superconducting thin films showed YBCO (00l) diffraction peaks. The strongest intensity was observed at 3000 Pa. Fig. 1 (b) shows the dependence of the c-axis orientation and T_c on the oxygen pressure. The highest T_c and c-axis orientation were obtained at around 3000 Pa.

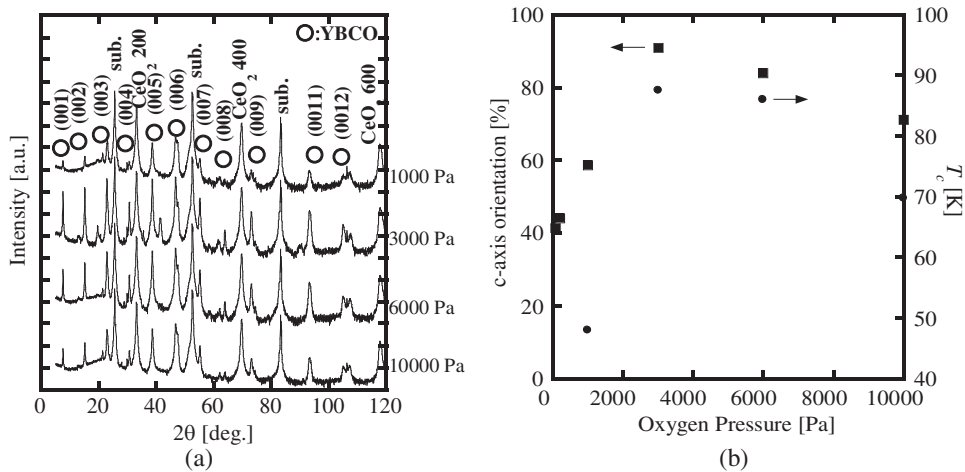


Fig. 2 (a) XRD-2θ/θ patterns varying the oxygen pressure.
(b) Dependence of the c-axis orientation and T_c on the oxygen pressure.

3.2. Crystallinity properties of thin films

The YBCO superconducting thin films were fabricated from amorphous YBCO thin films by using the optimal post-annealing conditions shown in Fig. 2. Fig. 3 (a) plots an X-ray diffraction (XRD) pattern of YBCO superconducting thin films and amorphous YBCO thin films. YBCO/CeO₂/r-Al₂O₃ superconducting thin films showed strong YBCO (00l) and CeO₂ (h00) diffraction peaks, but no YBCO (00l) diffraction peak of the amorphous YBCO thin film was observed. Thus, the observations of peaks of YBCO phase suggested that the YBCO/CeO₂ thin films grew c-axis orientation on the r-Al₂O₃ substrate. Using the Nelson-Riley (N-R) function, the c-axis length was estimated 11.67 Å.

Fig. 3 (b) shows X-ray ϕ -scan diffraction patterns of YBCO/CeO₂/r-Al₂O₃ thin films. We used the YBCO (102), CeO₂ (220), and r-Al₂O₃ (104) diffraction for the ϕ -scan measurements. Both YBCO and CeO₂ peaks showed four-fold symmetry and rotated 45 degrees between YBCO (102) grain axis and CeO₂ (220) grain axis.

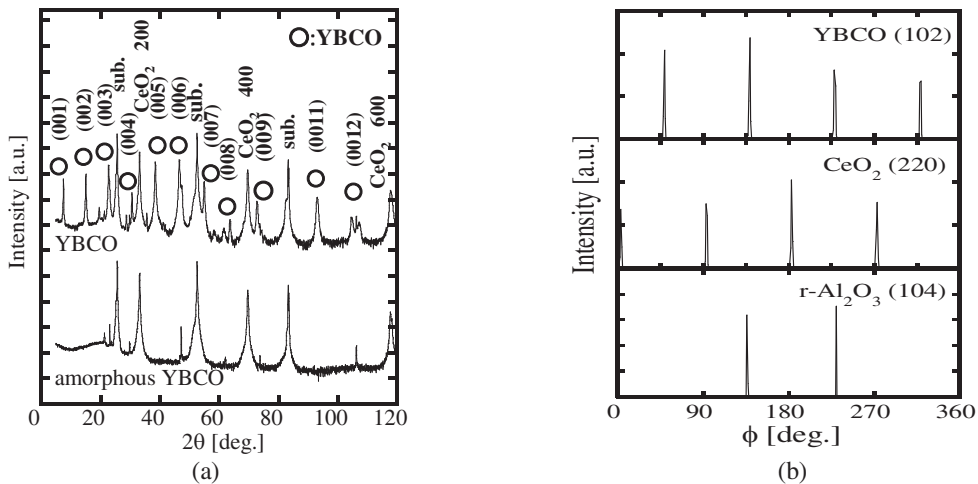


Fig. 3 (a) XRD-2θ/θ pattern of YBCO thin films and amorphous YBCO thin films.
(b) ϕ -scan of YBCO(102)/CeO₂(220)/r-Al₂O₃(104)

3.3. Superconducting properties

Fig. 4 (a) shows the normalized resistance vs. temperature of the YBCO thin films. The YBCO thin films showed the zero resistance at 87 K, and the residual resistance ratio (RRR = $R_{300\text{ K}}/R_{100\text{ K}}$) was 2.75. The produced YBCO thin films showed metallic behavior above T_c .

Fig. 4 (b) shows temperature dependence of microwave surface resistance (R_s) of the YBCO thin films. We measured the R_s using the dielectric resonator method. The resonance frequency of the TE011 mode was set to 21.8 GHz. The R_s value was about 2 mΩ at 21.8 GHz and at 20 K. The R_s of YBCO thin films were approximately one order of

magnitude smaller than that of the copper metals, which are usually used for microwaves. The R_s of our YBCO/CeO₂/r-Al₂O₃ thin films suited the filter application. We found that the optimal post-annealing conditions for fabricating the superconducting thin films with good crystallinity, T_c of 87 K, and low R_s from amorphous YBCO thin films. These results suggest that, we can fabricate high-quality HTS device starting from YBCO amorphous films, since the oxygen deficit at the edge of the patterns by the etching may be able to decrease. Additionally, this process has a possibility of fabricating a miniaturized pattern of the HTS thin films.

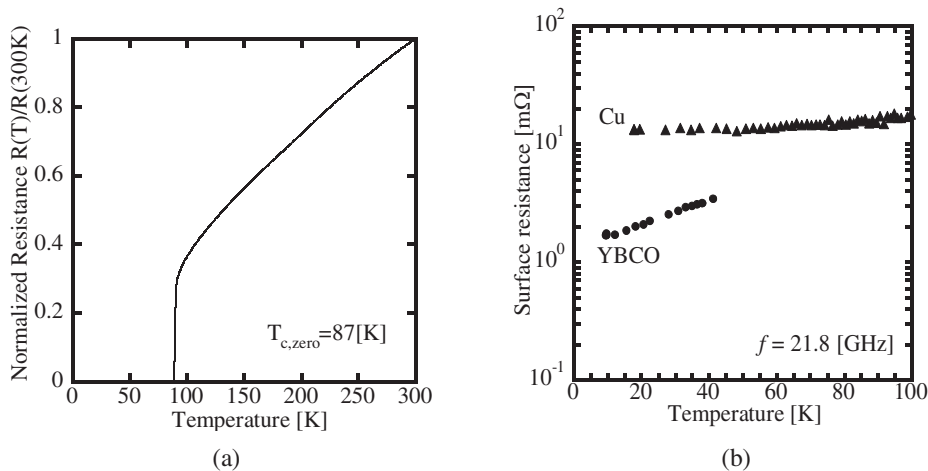


Fig. 4 (a) Normalized resistance vs. temperature of YBCO thin films.
(b) Temperature dependence of R_s .

4. Conclusion

We examined the phase, crystal grain orientation, and superconducting critical temperature of YBCO thin films formed from amorphous YBCO thin films by using a post-annealing method. The YBCO thin films were oriented epitaxially on a CeO₂/r-Al₂O₃ substrate and had a T_c of about 87 K. Therefore, our process was useful to make YBCO microstrip line devices. We are currently studying how to make a filter and resonator by using YBCO microstrip lines made by this process.

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References

- [1] M. K. Wu, et al. Phys. Rev. Lett., 58, 908, (1987).
- [2] H. Sato, et al. Jpn. J. Appl. Phys., 32, 1044, (1992).
- [3] S. K. Mishra, et al. Journal of Supercond., Vol.5, No. 5, 445, (1992).
- [4] H. Akoh, et al. IEEE Trans, Vol. 3, No. 1, (1993).
- [5] M. Matsumoto, et al. J. Appl. Phys., 66, 3907-3909, (1989).
- [6] T. Genichi, et al. IEICE, C-401, 51, (1996).
- [7] J. Sato, et al. IEICE, J93-C, 311, (2010).
- [8] M.W. Denhoff, MacCaffrey, J. Appl. Phys., 3986, (1991).
- [9] A.G. Zaitsev, R. Wondenweber, G. Ockenfuss, R. Kutzner, T. Konigs, C. Zuccaro, N. Klein, IEEE Trans. Appl. Supercond., 1482, (1997).
- [10] S. Ohshima, et al., Physica C., 335, 207–213, (2000).
- [11] S. Ohshima, et al., JCSJ., Vol. 39, No. 8, 354-358, (2004).